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**Measurements of Atomic Interactions and Frequency
Stability with a Cryogenic Hydrogen Maser**

AFOSR Grant 91-0282

Final Report

For the period 11 June 1991 through 30 June 1993

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MEASUREMENTS OF ATOMIC INTERACTIONS AND FREQUENCY STABILITY WITH A CRYOGENIC HYDROGEN MASER

ABSTRACT

With support from the Air Force Office of Scientific Research, we have developed a hydrogen maser that operates near 0.5 K using a high-cooling-power ^3He refrigerator. We have used this cryogenic hydrogen maser to study low temperature atomic hydrogen spin-exchange collisions and hydrogen-helium interactions, and to investigate the performance of cryogenic hydrogen masers as possibly improved frequency standards.

INTRODUCTION

The atomic hydrogen (H) maser is the most stable time and frequency standard currently available for measurement intervals between 10 and 10^5 seconds. The frequency stability of state-of-the-art H masers, such as those designed and constructed by our group at the Harvard-Smithsonian Center for Astrophysics, is typically better than one part in 10^{15} for averaging intervals of 10^4 seconds. The H maser is an essential frequency reference for radio astronomy, time-keeping, and spacecraft navigation, and is used in precision tests of gravitation, relativity, and quantum mechanics. In addition, it is a valuable instrument for experimental atomic physics, including work at low temperatures.

A cryogenic hydrogen maser (CHM) operating at temperatures below 1 K is a valuable experimental tool because it has the potential to provide substantially improved frequency stability, and because it enables the study of newly identified effects in ultra-low energy atomic collisions.

A CHM could have a frequency stability that is two to three orders of magnitude better than a room temperature H maser due to: (i) lower thermal noise; and (ii) a reduced H-H spin-exchange line-broadening and the resulting higher operational signal power. In 1986 three groups [1,2,3], including ours,

demonstrated oscillation of a CHM using superfluid helium-lined containers. It has subsequently been predicted by the Eindhoven group [4,5] that new low temperature H-H spin-exchange effects may make it difficult to achieve a frequency stability better than one part in 10^{16} using a CHM. In practice, the magnitude of these collisional effects and the ultimate frequency stability that can be realized by the CHM remain to be determined.

A CHM with significantly improved frequency stability would have many important scientific and technical applications. In particular **the CHM is the only active oscillator under development with the potential for short-term frequency stability of one part in 10^{16} or better.** Such active oscillator stability will be required for use with proposed "next generation" time and frequency standards utilizing stored ions or atomic fountains.

The experimental investigation of low temperature H-H collisions is scientifically interesting because the spin-exchange frequency shift and line-broadening cross-sections depend sensitively upon (i) details of the H-H interaction potential at long range that are otherwise experimentally inaccessible; (ii) non-adiabatic (i.e. non-Born-Oppenheimer) effects unique to cold atomic collisions; and (iii) inclusion, in addition to the electron exchange interaction (strength $\sim 50,000$ K), of the intra-atomic hyperfine interaction (strength ~ 0.07 K), which fundamentally alters the rotational symmetry of the H-H collisional process. Furthermore, the calculated values for these spin-exchange cross-sections have been found to be sensitive to the theoretical techniques used in the calculations [4,5,6], making experimental comparisons particularly significant. Similar low temperature spin-exchange phenomena can occur with heavier alkali atoms: theoretical analyses have been performed by the Eindhoven group for Cs-Cs [7,8] and Na-Na [9] collisions, and recent measurements of Cs-Cs spin-exchange effects have been reported by Gibble and Chu [10] using a laser-cooled atomic fountain; these measurements and calculations for Cs disagree significantly with one another. A comparison of H-H spin-exchange calculations with measurements from the CHM will provide a better test of theory than in the heavier alkali atoms because the interaction potential of the H-H system is simpler and better understood.

HARVARD-SMITHSONIAN CHM

The current Harvard-Smithsonian CHM research program was begun in June, 1991 with support from the Air Force Office of Scientific Research (AFOSR). As a part of that program, we have developed a high cooling-power, recirculating ^3He refrigerator dedicated to CHM use. This refrigerator will provide 2 mW of cooling power at 0.5 K when operated in the recirculating mode in tandem with the CHM. A detailed discussion of the design and operation of the Harvard-Smithsonian CHM has been published previously [3]. In the CHM, H atoms from a room temperature microwave discharge enter the ^3He refrigerator and are cooled to ~ 10 K. The atomic beam passes through a hexapole magnet that focusses the two upper hyperfine states into a microwave cavity resonant at 1420 MHz that is maintained at ~ 0.5 K in a region of stable and homogeneous magnetic field. The cold H atoms are stored within the cavity for times ≥ 2 seconds in a chamber coated with a superfluid helium film. With a modest input H flux ($\geq 10^8 \text{ sec}^{-1}$) the Harvard-Smithsonian CHM achieves continuous maser oscillation. The CHM signal is detected inductively, isolated, conducted out of the ^3He refrigerator, and then amplified. The frequency and power of this output signal are precisely measured. Room temperature H masers in our laboratory serve as frequency comparison standards for the CHM.

We can independently control the input H flux to the CHM, the resonant cavity frequency, the magnitude and profile of the static magnetic field, and the system temperature. We can also apply audio frequency radiation to the H ensemble stored in the CHM cavity to drive Zeeman transitions among the upper three hyperfine states. As discussed in our original CHM proposal to the AFOSR (dated 11 April 1993), these capabilities allow us to measure low temperature H-H spin-exchange frequency shift and line-broadening effects with the CHM.

EXPERIMENTAL RESULTS

In this section we describe our experimental results with the CHM during the period of the present AFOSR grant.

The complete Harvard-Smithsonian CHM/ ^3He refrigerator system has been operational since early 1993. During this period the condensing impedance failed in the refrigerator's ^3He return line. This meant that the refrigerator had to

be operated in a batch, or one-shot, mode, giving approximately three hours of running time per day rather than continuous operation. Much of this three hour run time is needed for cooling the CHM to ~ 0.5 K and stabilizing the maser oscillation. Thus the effective data-taking time was typically less than 30 minutes per day.

We have operated the CHM with a maximum H input flux $\approx 10^{12} \text{ sec}^{-1}$, resulting in a maximum signal power $\approx 10^{-12}$ watts and a maximum stored H density $\approx 10^{10} \text{ cm}^{-3}$. We have measured the CHM output frequency as a function of temperature (see Fig. 1). The CHM signal frequency is decreased by H collisions both with the He-film-coated walls of the storage chamber and with the accompanying He vapor. These two negative frequency shifts have opposite trends with temperature, since the H-wall residency time decreases with temperature and the He vapor pressure increases with temperature. Thus there is a stable minimum in the magnitude of the total H-He frequency shift. For the current CHM geometry this minimum is near 0.53 K (see Fig. 1).

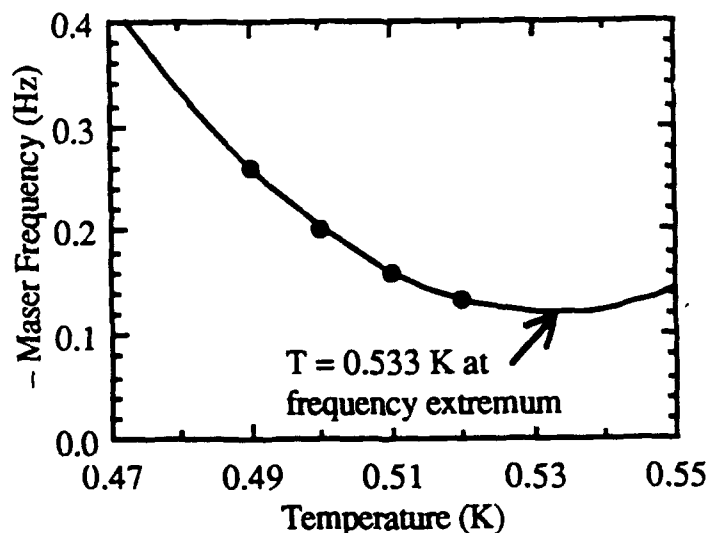


Figure 1. Measured dependence of CHM signal frequency on temperature, and a quadratic fit to data.

Our measurements show a stronger dependence of CHM frequency on temperature than that reported previously by the University of British Columbia (UBC) group using a different experimental technique [11]. We believe that this larger "wall shift" in our system is due to H interactions with paramagnetic impurities in the sapphire substrate that lies beneath the He film. (Note that

there is also a 50 μm layer of Teflon between the sapphire and the He.) The hypothesis of strong H-wall interactions in our CHM is supported by two other measurements:

- 1) We have determined the absolute value of the H-wall frequency shift = $-0.297(5)$ Hz at $T = 0.490(2)$ mK. The results of the UBC group, however, imply a much smaller frequency shift ≈ -0.1 Hz.
- 2) We have measured our CHM's line-Q $\approx 2.3 \times 10^9$ with insignificant magnetic and collisional relaxation, implying $T_2 \approx 0.52$ seconds. The contribution to T_2 from H escape from the storage bulb is calculated from geometry to be approximately 2.5 seconds. Thus we conclude that the observed T_2 is primarily due to wall relaxation.

We have also measured the CHM's frequency stability relative to a room temperature H maser. At $T = 0.49$ K, with 20 μK temperature control and a signal power $\approx 10^{-14}$ W, we found $\sigma_y(\tau) \approx 1.6 \times 10^{-13}/\sqrt{\tau}$, for 10 seconds $< \tau < 160$ seconds. Here $\sigma_y(\tau)$ is the Allan, or two-sample, deviation [12], a measure of fractional frequency stability over an averaging interval τ . (The upper limit on τ is due to the limited data-taking time we have had without recirculating refrigerator operation.) A typical set of data comparing CHM and room temperature H maser frequency stability is shown in Fig. 2. Also shown in Fig. 2

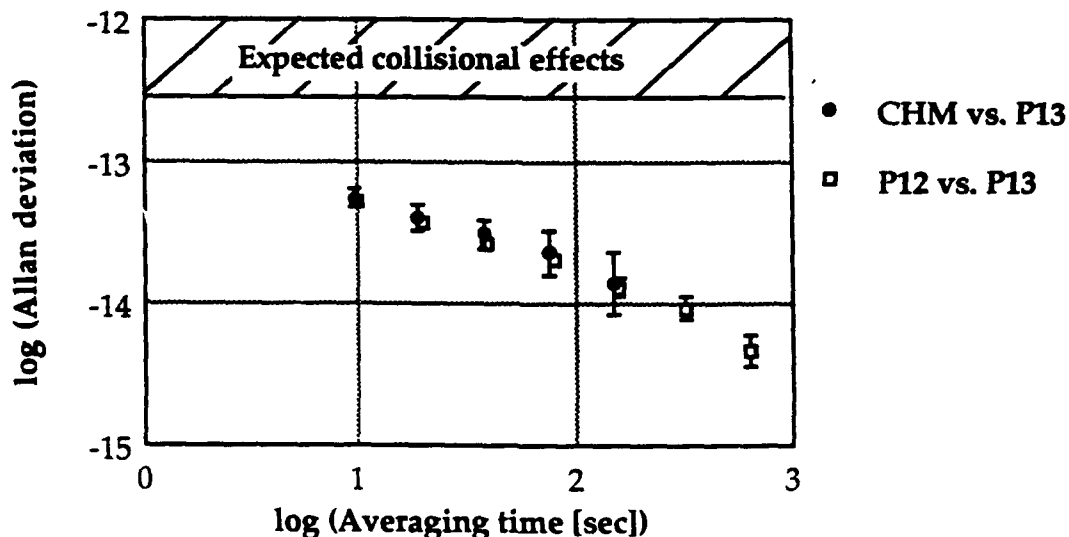


Figure 2. Typical measured frequency stabilities of the CHM relative to a room temperature H maser (P13), and P13 relative to a second room temperature H maser (P12).

is the measured stability of the reference H maser (designated P13) relative to a second room temperature H maser (P12). It is clear from this data that the determination of the CHM's short-term frequency stability is already limited by the performance of the room temperature H masers.

Finally, we have made preliminary measurements of H-density-dependent spin-exchange line-broadening in the CHM. An example of such a measurement is shown in Fig. 3. (The effect of the limited data-taking time is evident in that only two-point data sets have been made.) Here the CHM's microwave cavity was tuned ~ 10 kHz off-resonance, so that the observed density-dependent frequency shift gives information on the spin-exchange line-broadening cross section $\bar{\sigma}_{se}$. From several two-point data sets at $T = 0.49$ K, such as that shown in Fig. 3, we find $\bar{\sigma}_{se} = 2\text{--}3 \times 10^{-16} \text{ cm}^2$, whereas recent calculations [5] predict $\bar{\sigma}_{se} \approx 8 \times 10^{-17} \text{ cm}^2$. Note that an absolute H density calibration has not been completed for these preliminary CHM measurements. The H densities listed in Fig. 3 are lower limits; higher H densities would bring the experimental and theoretical values for $\bar{\sigma}_{se}$ into better agreement. Nevertheless, these preliminary H-H collision measurements demonstrate the efficacy of the CHM as an experimental tool for studying low temperature atomic physics.

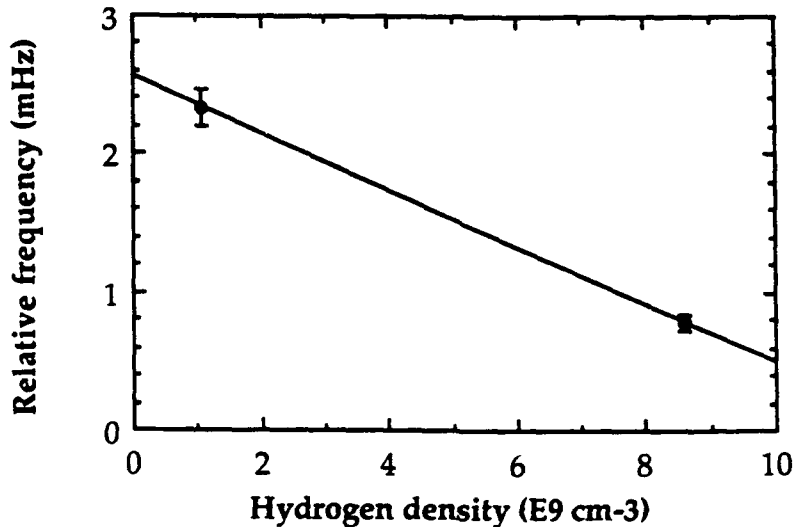


Figure 3. Example of preliminary measurements of H-density-dependent frequency shifts in the CHM.

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